Measurements of the Aerosol Light-Scattering Coefficient at Ambient and 85% Relative Humidity on the ONR Pelican During ACE-2

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LONG-TERM GOALS

The ultimate goal of this study, as originally conceived, is the determination of aerosol hygroscopicity and its impact on physical (e.g., size) and optical (e.g., scattering coefficient) properties of marine aerosols on large spatial scales. In the course of our analysis, it has further become clear that relating aerosol hygroscopicity to chemical composition, particularly organic composition, is an integral component of this research. Additionally, associated with the renewal of funding in 2002, goals concerning the interaction of marine aerosols with marine boundary layer (MBL) clouds have been added. These new goals include: 1) assessment of the relative impacts of purely meteorological and aerosol factors on cloud albedo, 2) determination of the impact of very large CCN on cloud albedo, 3) assessment of the impact of cloud processing on aerosol light scattering, and 4) explore, observationally, the impact of organics on CCN activation in MBL clouds.

OBJECTIVES

Several of the objectives enumerated in previous reports have now been largely achieved (e.g., assessment of the impact of humidity on aerosol scattering coefficient for different aerosol types in ACE-2, remote retrieval of aerosol hygroscopicity and CCN concentration). Hence, in this report, we move on to objectives which have yet to be achieved, or have been achieved in the last year, and emphasize the more recent data sets which will be utilized to address them. For example, results from the ACE-Asia and RED experiments are now available. Furthermore, analyses of data gathered during the recent CALSPAN laboratory experiment (ONR sponsored) are also available to help guide the current analyses. Finally,while preliminary results from the CARMA study, conducted to address the new objectives alluded to above, are just available, detailed discussion of these results would be premature at this point. Hence, our objectives are as follows:

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14. ABSTRACT
The ultimate goal of this study, as originally conceived, is the determination of aerosol hygroscopicity and its impact on physical (e.g., size) and optical (e.g., scattering coefficient) properties of marine aerosols on large spatial scales. In the course of our analysis, it has further become clear that relating aerosol hygroscopicity to chemical composition, particularly organic composition, is an integral component of this research. Additionally, associated with the renewal of funding in 2002, goals concerning the interaction of marine aerosols with marine boundary layer (MBL) clouds have been added. These new goals include: 1) assessment of the relative impacts of purely meteorological and aerosol factors on cloud albedo, 2) determination of the impact of very large CCN on cloud albedo, 3) assessment of the impact of cloud processing on aerosol light scattering, and 4) explore, observationally, the impact of organics on CCN activation in MBL clouds.

15. SUBJECT TERMS

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- Explore the relationship between aerosol hygroscopicity and organic composition (ACE-Asia, RED, CARMA, possibly ACE-2).

- Further exercise of the remote retrieval techniques for hygroscopicity and CCN concentration described previously to new data gathered during CARMA.

- Explore the impact of aerosols on cloud albedo and the impact of cloud processing on aerosol properties.

APPROACH

The technique used to quantify the hygroscopicity of aerosols from in-situ measurements has been described in previous reports. Similarly, the basic approach used to retrieve aerosol hygroscopicity from remote sensing measurements, namely, the use of a Look-Up Table (LUT) based on that employed by the MODIS team has been mentioned previously.

Concurrent analyses of the chemical composition of the aerosol, and in particular the organic composition, are now routinely done. Aerosol samples are first obtained by either filtration or impaction (filter packs or MOUDI impactors) and subjected to an array of analytical techniques upon return to the laboratory. These include ion chromatography with both conductivity and pulsed amperometric detectors, and LC-MS. Relating the analyzed aerosol composition to the hygroscopicity is being undertaken on both an empirical basis via multiple regression and factor analysis, and more fundamentally utilizing a variant of the Ming/Russell aerosol model (Ming and Russell, 2001).

Methodologies for analysis of the data relevant to the third objective – aerosol/cloud interactions – will be partially dependent on the type and quality of data acquired (which is still being assessed) but will no doubt utilize both diagnostic modeling and statistical analysis.

WORK COMPLETED

Tasks associated with the second of the three objectives listed above have essentially been completed utilizing data from the TARFOX and ACE-2 data sets. A manuscript reporting these results is now in press (Gasso and Hegg, 2002). However, we are further exploring the feasibility of applying the techniques developed to the ACE-Asia and CARMA data sets. Data from ACE-Asia on the first of the above objectives have also been analyzed and a manuscript submitted for publication. Data on the third objective have in fact been obtained during the just completed CARMA study but no analysis has as yet been done.

RESULTS

Preliminary to the investigation of the relationship between hygroscopicity and chemical composition, we felt it necessary to expand our analytical capabilities to permit a more complete speciation of the ambient aerosol mass. To this end we expanded our suite of analyses to include IC-PAD and LC-MS. This has permitted the identification of carbohydrates as well as the more readily identified carboxylic acids. We first field tested our expanded suite by participation in the SAFARI-2000 field campaign. Figure 1 shows the results from the analysis of 33 filter samples obtained in regional haze during the field campaign. While the speciated mass fractions still fall well short of unity, they are commonly much higher than the typical 40% speciated mass reported in previous studies.

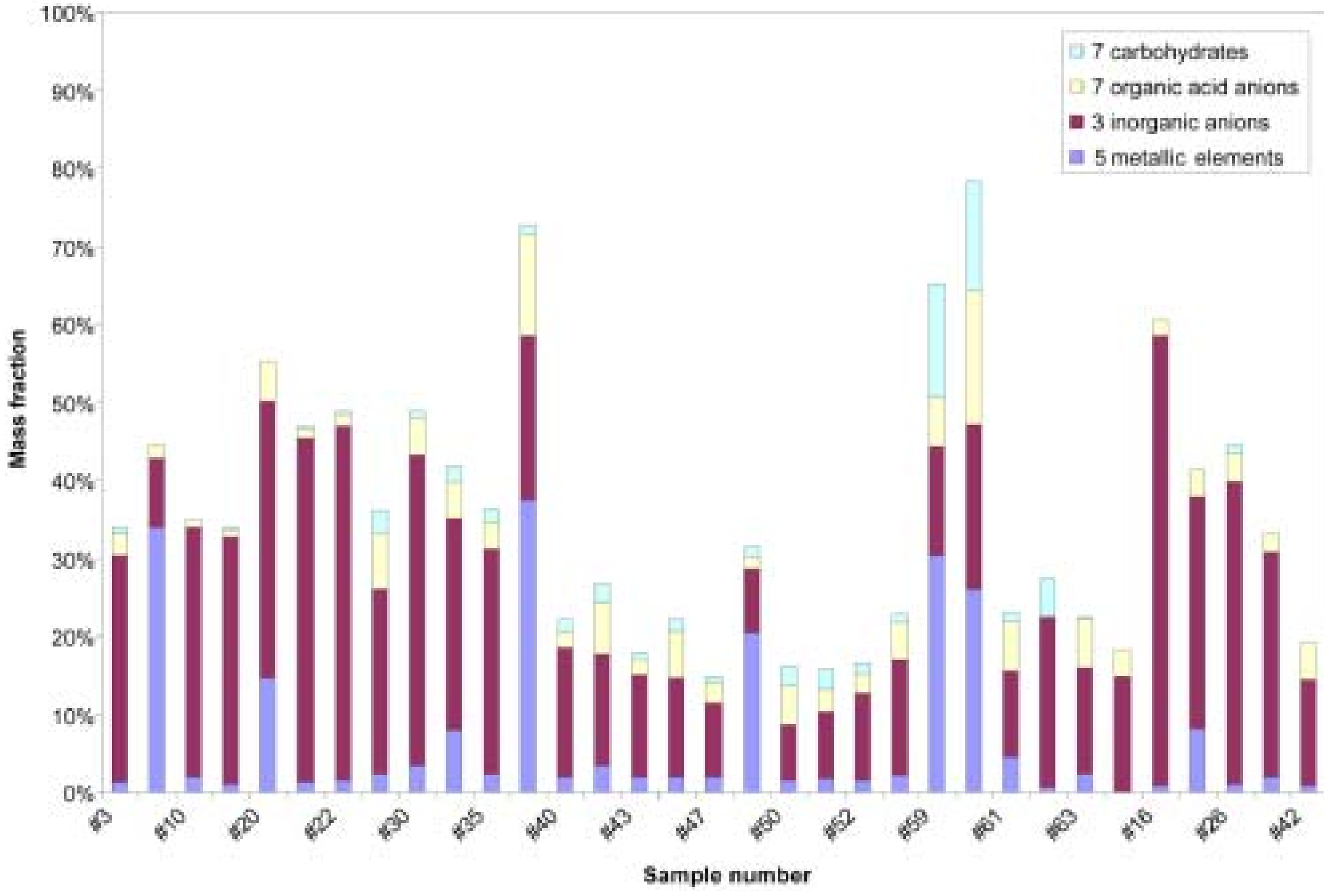

Figure 1 Fractional contributions of four aerosol classes of chemical species (carbohydrates, carboxylic acids, inorganic anions and metallic elements) to the total mass of haze aerosols measured on 33 filter samples in SAFARI 2000

The same sort of analyses have recently been performed on filter samples obtained during the ACE-Asia campaign. It was found that the organic component of the aerosol was appreciably lower in the ACE-Asia data than was the case in SAFARI but, interestingly enough, factor analysis suggested that there was a substantial biomass burning component to the aerosol light scattering. Indeed, it appears that the aerosol has four major components/sources with respect to aerosol hygroscopicity and that one of these is biomass burning (the others being dust, pollution and soluble organics). In Figure 2, we compare results from an empirical model of aerosol hygroscopicity, using literature values for the hygroscopicity of the various aerosol components (in terms of the γ parameter previously discussed) coupled with their mass fractions, with actual measurements of γ derived from our humidigraph data (again as previously discussed). The agreements is quite reasonable in our view and suggests that organics, either from biomass burning or other sources, are a major modulator of aerosol hygroscopicity in the important ACE-Asia venue.

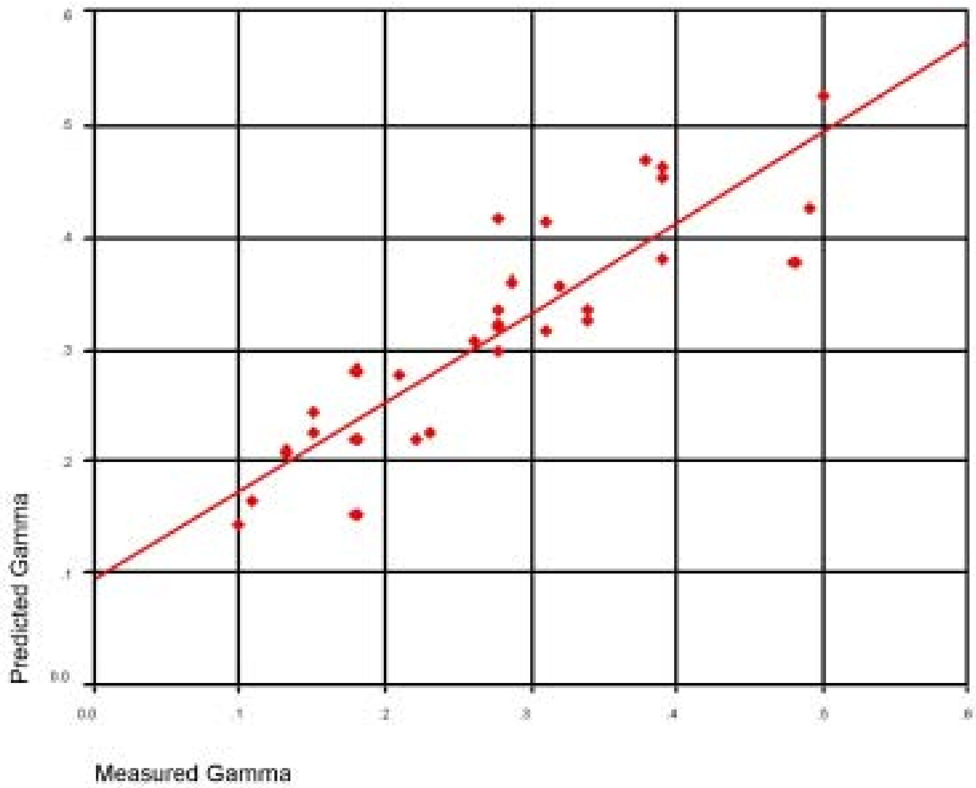

Figure 2 Comparison of values of γ derived from an empirical model with those calculated from humidigraph data obtained during ACE-Asia.

More generally, hygroscopicity in marine air has been evaluated by comparison of values of γ obtained from three separate field campaigns in the Pacific (DECS, ACE-Asia and RED). The values obtained are relatively low, even in background air, consistent once again with a substantial organic component.

IMPACT/APPLICATIONS

These results suggest that organics commonly play an important role in the hygroscopicity of marine aerosols. Hence, to properly assess, and predict, the impact of RH on visibility in the MBL, this organic aerosol component must be taken into account. Furthermore, the nature of the organic component, i.e., its speciation, must also be understood. Unlike the traditional inorganic sea salt model, in which the speciation is not of paramount importance since nearly all inorganic salts have roughly similar hygroscopicities, different organics can have quite different impacts upon aerosol hydration (e.g., Ming and Russell, 2001; Ellison et al, 1999).

TRANSITIONS

For the current reporting period, several different data sets have been acquired and should yield interesting information. These data sets, ACE-Asia, RED and CARMA, contain data on both hygroscopicity and chemical composition. Such data are of wide interest to the aerosol community.

RELATED PROJECTS

The use of aerosol hygroscopicity and associated optical data (e.g., scattering and absorption coefficients for ambient aerosols) is of great interest to various research groups. For example, we are currently working with Phil Russell's group at NASA-Ames in a closure study of aerosol optical depth based on the ACE-Asia data set. A manuscript based on this work will shortly be submitted for publication. Similarly, retrieval of both aerosol hygroscopicity and CCN concentration are of great interest to the remote sensing community. We are exploring the feasibility of in-situ/remote retrieval comparisons of both hygroscopicity and CCN concentration with data from both the ACE-Asia and CARMA studies. Finally, we are working with Carl Friehe and his colleagues at UC Irvine, and Phil Durkee and his colleagues at NPS on the recently acquired CARMA data set. Our goals are to better understand the roles of both meteorological and aerosol parameters on MBL cloud albedo and longevity.

SUMMARY

Our work over the last year has lead to two conclusions which constitute advances in our knowledge relative to a year ago.

- Aerosol hygroscopicity in the marine atmosphere is commonly much less than that predicted based on traditional inorganic sea salt composition. Organics appear to play a key role in the hygroscopicity of marine aerosols.

- Aerosol light scattering and hygroscopicity in the ACE-Asia study area are due to a handful of aerosol components and thus sources, pollution, biomass burning and dust being the important players.

In the next few years, we hope to relate measured aerosol composition with concurrently measured aerosol hygroscopicity and optical properties via a more firmly based theoretical model, essentially a varient of the Ming/Russell model. With this, we hope to assess how changes in the aerosol composition due to in-situ chemical reaction effects the aerosol hygroscopicity and associated optical properties. And , of course, with the new CARMA initiative, we will be assessing the roles of meteorological and aerosol parameters on MBL cloud albedo.

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PATENTS

None.